Spectroscopic Evaluation of Yb^{3+}-Doped Glasses for Optical Refrigeration

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Abstract—**The absorption and emission properties of Yb^{3+}-doped ZBLANP, BIGaZYT and QX/Yb phosphate glasses are studied to evaluate their potential for laser-induced fluorescent cooling or optical refrigeration. The efficiency of optical refrigeration increases with pump wavelength in the anti-Stokes region. The cooling efficiencies of the three glasses as a function of temperature are evaluated at the wavelength λ_p corresponding to the absorption coefficient of 10^{-3} cm^{-1}. For temperatures <110 K, the cooling efficiency of the BIGaZYT glass may be more than twice that of the ZBLANP.**

Index Terms—**Laser cooling, optical properties of solids, optical refrigeration, rare-earth doped glasses.**

I. Introduction

THE CONCEPT of cooling an object through its interaction with monochromatic radiation was proposed in 1929 by Pringsheim [1]. After Landau [2] established the basic thermodynamic consistency of such a process, aspects of fluorescent cooling were vigorously pursued [3]–[9]. Much progress has been achieved in laser cooling free atoms to far below one microkelvin [10], [13]–[16]. In contrast, attempts to cool condensed phase material with light have met with limited success [5]. Optical materials such as GaAs [17] and Nd:YAG [5] were proposed as candidates for solid-state optical refrigerators, but the first steady-state optical refrigeration was demonstrated with a ytterbium-doped fluoride glass, Yb:ZBLANP [18]. In subsequent experiments a 16 K temperature decrease was achieved using an optical fiber of this material [19].

The basic condition for optical refrigeration is that a material exhibit high-quantum-efficiency anti-Stokes fluorescence. A material emits photons of greater mean energy than those absorbed, and the energy difference is supplied by thermal phonons. This situation is realized by pumping the cooling material with monochromatic radiation with a wavelength λ_p longer than the wavelength $\overline{\lambda}$ that corresponds to the mean fluorescence photon energy. This is similar to running a four-level optically pumped laser backward, i.e., pumping at the "lasing" wavelength and emitting at a "pumping" wavelength, as depicted in Fig. 1. The pump laser is tuned such that the

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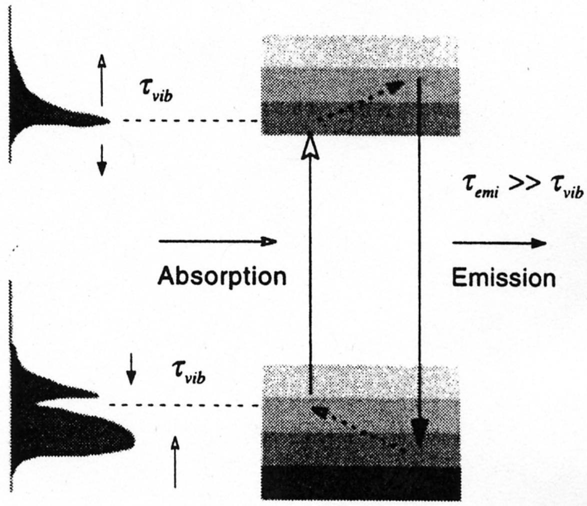

Fig. 1. Schematic illustration of laser-induced fluorescence cooling, where τ_{em} is the radiative lifetime and τ_{vib} is the phonon relaxation time.

absorption occurs from near the top of the lower manifold to near the bottom of the upper one. A short-lived "hole" is created at top of the lower manifold and a "peak" at the bottom of the upper manifold. The reequilibrating processes, i.e., the refilling of the hole and spreading of the peak, lead to phonon absorption and cooling of glasses.

For constructing a practical cryocooler, additional physical properties are desirable. For example, a large dopant concentration permits the cooling element to be compact. High thermal conductivity decreases the temperature drop within the cooler. Radiation resistant materials are important for space applications, and mechanical durability enhances the cooler's ruggedness. Reference [20] describes the design and performance of optical refrigerators. These devices have the potential advantages over mechanical cryocoolers of no vibrations, long lifetimes, and no electromagnetic interference.

The Yb^{3+}-doped glasses feature an energy scheme similar to that pictured in Fig. 1. The trivalent ytterbium has only two available manifolds, the $^2F_{7/2}$ ground state and $^2F_{5/2}$ excited state, separated by $\sim$10 000 cm^{-1}, with moderately strong electric-dipole transitions, and relatively strong electron–phonon coupling strength. In particular, the lack of three unwanted effects: multiphonon relaxation, concentration quenching, and excited state absorption make them excellent candidates for laser-induced fluorescent cooling. The closely spaced energy levels of Yb^{3+} ions in the inhomogeneous

TABLE I
PHYSICAL AND THERMAL PROPERTIES OF THE ZBLANP, BIGAZYT, AND QX PHOSPHATE GLASSES

Properties	units	ZBLANP	BIGaZYT	QX/Yb
Density	g/cm^3	4.414	5.44	2.81
Refractive index	n	1.49	1.51	1.53
Yb^{3+} site density	$N(10^{20}cm^{-3})$	1.128	19.68	2.215
Glass transition temp.	^0C	256	330	450
Crystallization temp.	^0C	331	442	-
Thermal expansion	$10^{-7}K^{-1}$	200	150	83
Temperature coef.	$dn/dT(10^{-5}K^{-1})$	-1	-1.2	-0.1
Heat capacity	J/gK	0.575	0.418	-
Thermal conductivity	W/mK	10^{-3}	$10^{-3\sim4}$	0.85

environments of the glassy host enable operation at useful low temperatures where only low-energy phonons are available. The large energy gap of the trivalent ytterbium between the ground and first excited states, which is at least a factor of ten greater than the maximum phonon energy of the glasses, allows a high quantum-yield fluorescence; nonradiative decays from the upper manifold require simultaneous emission of many phonons and are therefore strongly inhibited.

The most fundamental spectroscopic requirement for optical refrigeration is that there exists significant absorption extending to longer wavelengths than the mean fluorescent wavelength $\overline{\lambda}$. In this paper, we present an assessment of the optical refrigeration potential of three materials, Yb^{3+}-doped BIGaZYT, QX phosphate, and ZBLANP glasses, based on measurements of absorption and emission spectra and their temperature dependencies. The reciprocity method has been used to obtain better absorption coefficients at the red tail where the wavelength is longer than the mean fluorescence. To estimate the practical cooling efficiencies of these materials as functions of temperature, we calculate the efficiencies at pumping wavelengths corresponding to an absorption coefficient of 10^{-3} cm^{-1}.

II. EXPERIMENTAL

The glass samples we investigated were the Yb^{3+}-doped ZBLANP, BIGaZYT, and QX phosphate glasses. The compositions are: 53ZrF$_4$, 18BaF$_2$, 3LaF$_3$, 3AlF$_3$, 20NaF, 2PbF$_2$ and 1YbF$_3$ for ZBLANP (1 wt%YbF$_3$); 30BaF$_2$, 18InF$_3$, 12GaF$_3$, 20ZnF$_2$, 6ThF$_4$, 4ZrF$_4$ and 10YbF$_3$ for BIGaZYT (10 mol%YbF$_3$); and QX/Yb phosphate glass has 5 wt% of Yb_2O_3. Table I is a compilation of general physical and thermal properties of the three host materials. The listed properties are important for designing and fabricating practical coolers. The emission measurements were made using a tunable CW Ti:sapphire laser (Spectra-Physics Model 3900 S) pumped with an Ar^+ laser (Coherent INNOVA 20) and has a spectral bandwidth $\leq$ 30 GHz. A sample was placed in a cryostat (Infrared Lab. HDL-8) and the temperature was measured with a calibrated silicon diode. A fused silica optical fiber bundle directs the fluorescence to the entrance slit of the monochromator, a Digikrm 240 (CVI Laser Co.) equipped with a CCD array detector (Santa Barbara ST-6). The emission spectra were corrected for the CCD response and the attenuation of the fused silica fiber. The emission spectra of the Yb^{3+}-doped glass systems have the same shape regardless of the pumping wavelength, because the thermal reequilibrium processes (~nanoseconds) is so much faster than the radiative lifetime (~milliseconds).

The reabsorption from the ground state of the $^2F_{7/2}$ manifold can adversely affect the fluorescence spectra. This effect is most pronounced in the BIGaZYT and QX/Yb phosphate glasses which have 10 mol% YbF$_3$ and 5 wt% Yb_2O_3, respectively. The reabsorption was reduced by using a small sample and pumping near the surface that is observed. The absorption measurements were made with a similar setup, a calibrated white-light source replacing the laser. The sample was placed in the cryostat mounted on a micrometer stage. The transmission I_t and reference I_0 spectra were taken by moving the dewar container slightly such that the white-light beam passes through the sample for transmission and past the sample for reference. This procedure minimizes any changes due to window reflection losses.

III. RESULTS AND ANALYSIS

A. Absorption and Emission Spectra

Fig. 2 shows the room-temperature absorption coefficient α (cm^{-1}) and the emission spectra for all three glasses. The vertical dashed line indicates the wavelength $\overline{\lambda}$ that corresponds to the mean fluorescent photon energy, i.e.,

$$\overline{\lambda} = \frac{\int \lambda \varepsilon(\lambda)\, d\lambda}{\int \varepsilon(\lambda)\, d\lambda} \tag{1}$$

where $\varepsilon(\lambda)$ is the emitted power per unit wavelength. Absorption at wavelengths longer than mean fluorescence wavelength $\overline{\lambda}$ is defined as the cooling tail, since optical cooling can occur when materials are pumped at those wavelengths. We measured the absorption and emission spectra from temperatures of ~10–300 K to assess the temperature dependence of the cooling tails and mean fluorescence wavelengths. Fig. 3 shows

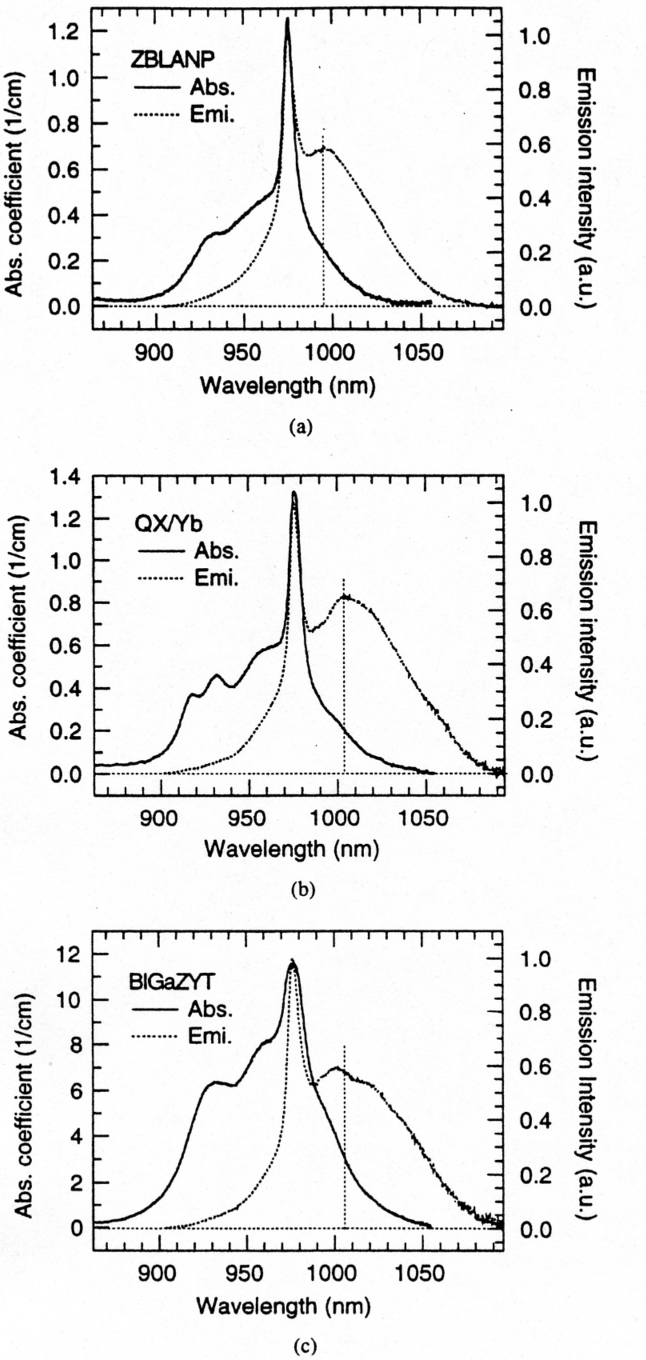

Fig. 2. The absorption and emission spectra at room temperature. The dashed vertical line indicates the mean fluorescence wavelength $\overline{\lambda}$. the absorption at wavelengths longer than $\overline{\lambda}$ is defined as the cooling tail. (a) $\overline{\lambda} \sim 995$ nm, (b) $\overline{\lambda} \sim 1004$ nm, and (c) $\overline{\lambda} \sim 1005$ nm.

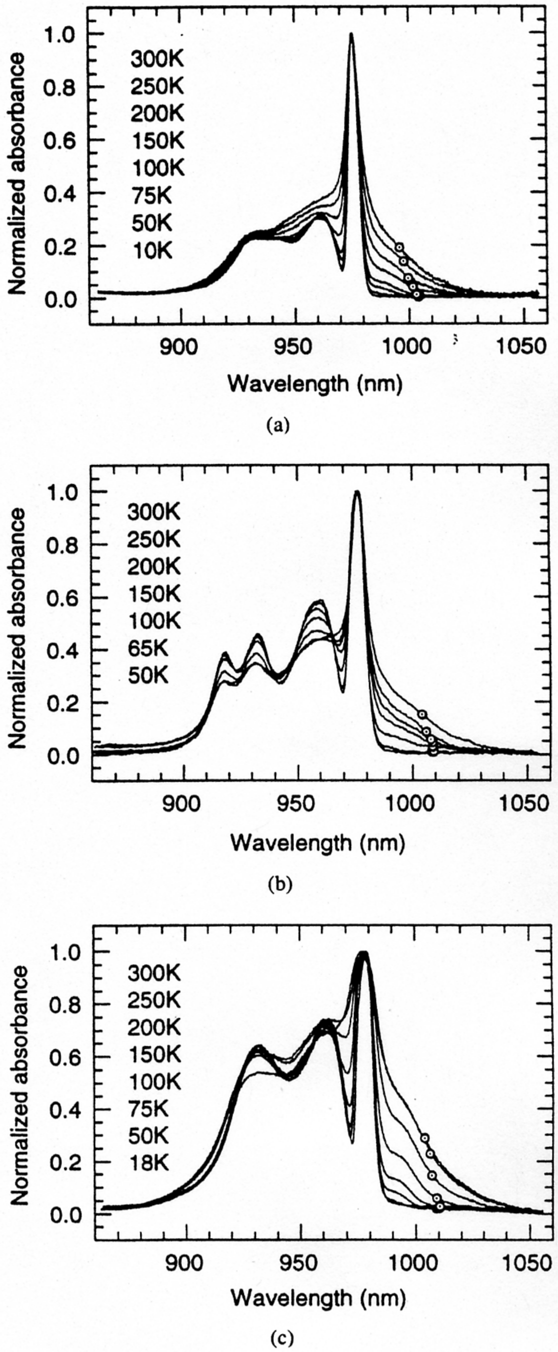

Fig. 3. The normalized absorption spectra at different temperatures for (a) ZBLANP, (b) QX/Yb, and (c) BIGaZYT. At long wavelengths, the cooling tail falls with decreasing temperatures. The open circles indicate the positions of the mean fluorescence wavelength.

the absorption spectra as a function of temperature, where the spectra were normalized at the peak of the $(^2F_{7/2})_1 \rightarrow (^2F_{5/2})_1$ transition near 975 nm; the open circles indicate the position of the mean fluorescence. The long-wavelength tail, the cooling tail, shows a strong temperature dependence; the absorption coefficient rapidly falls with decreasing temperature. Fig. 4 gives the temperature-dependent emission spectra. In this case, the short-wavelength blue tail of the emission decreases at lower temperatures. To determine the energy level structure of Yb^{3+} in these glasses, we analyzed the low-temperature (~10 K) absorption and emission spectra. We used three Gaussian line shapes to fit the $(^2F_{7/2})_1 \rightarrow (^2F_{5/2})_{1,2,3}$ transitions in absorption to determine the Stark-level positions in upper $^2F_{5/2}$; and four Gaussian line shapes to fit the $(^2F_{5/2})_1 \rightarrow (^2F_{7/2})_{1,2,3,4}$ transitions in emission for the lower $^2F_{7/2}$ manifold. Fig. 5 shows the energy level diagram of Yb^{3+} in ZBLANP, BIGaZYT, and QX/Yb glasses and the major transitions. A detailed spectral linewidth study of the temperature-dependent spectral behavior of ZBLANP:Yb^{3+} found that the homogeneous contribution to the width of the $(^2F_{7/2})_1 \rightarrow (^2F_{5/2})_1$ transition in the emission and absorption

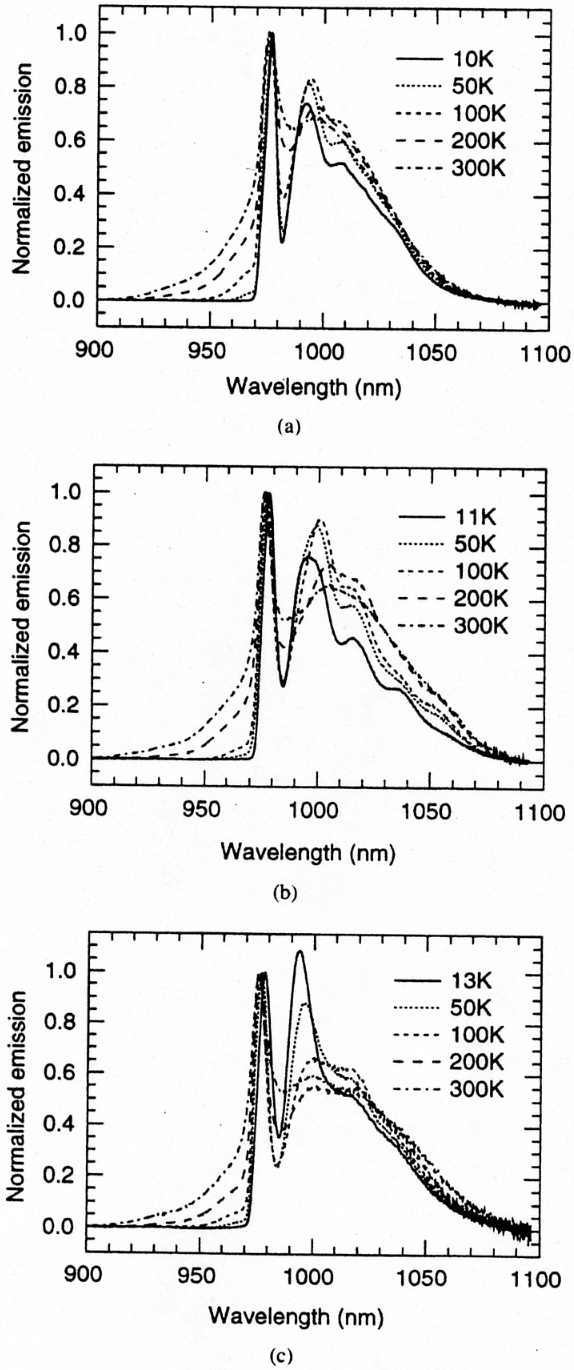

Fig. 4. The normalized emission spectra at different temperatures for (a) ZBLANP, (b) QX/Yb, and (c) BIGaZYT. The short-wavelength tail falls with decreasing temperature.

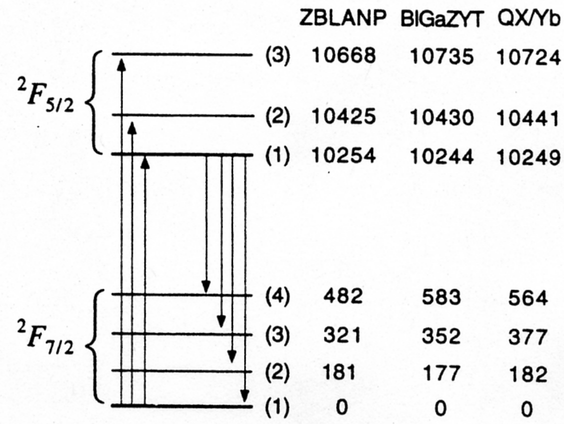

Fig. 5. Energy levels in cm^{-1} and major transitions of Yb^{3+} in ZBLANP, BIGaZYT, and QX/Yb glasses.

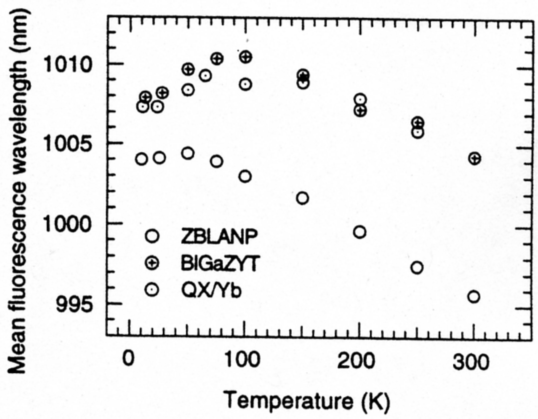

Fig. 6. Temperature dependence of the mean fluorescence wavelength of the ZBLANP, BIGaZYT, and QX/Yb phosphate glasses.

decreased approximately as T^2 for temperatures $10 < T < 300$ K [21].

The mean fluorescence wavelength $\overline{\lambda}$ for each of the emission spectra are plotted in Fig. 6 and indicated as dots on the absorption spectra of Fig. 3. Pumping the glass samples in the absorption tail to the right of the dots produces net anti-Stokes fluorescence and possible optical refrigeration. Because the population of the levels in the upper manifold follow a Boltzmann distribution, the mean fluorescent wavelength tends to increase with decreasing temperature, as is observed in Fig. 6 for temperatures above ~100 K. As the temperature falls, the fluorescence is increasingly due to decays from the $(^2F_{5/2})_1$ state, the lowest energy level in the upper manifold. Reabsorption inhibits escape of the fluorescent radiation corresponding to transitions to the $(^2F_{7/2})_1$ level, and to a lesser extent to the next lowest $(^2F_{7/2})_2$ level. These reabsorption effects, that are most pronounced in the BIGaZYT and QX/Yb phosphate glasses which have the highest Yb concentrations and the greatest absorption coefficients (see Fig. 2), increase at lower temperatures as the population of the lower energy Stark levels in the lower manifold grow. At the very lowest temperatures, below ~100 K, the population of the $(^2F_{7/2})_2$ level drops, decreasing the absorption for the corresponding radiation. One can see in Fig. 4 that the peak (~993 nm) of the $(^2F_{5/2})_1 \rightarrow (^2F_{7/2})_2$ transition in BIGaZYT grows with decreasing temperature below 100 K. The growth of this peak, which shifts $\overline{\lambda}$ to shorter wavelengths at the lowest temperatures, may be the result of declining reabsorption from the $(^2F_{7/2})_2$ level.

B. Reciprocity Method

To compare the potential cooling of the three glasses being examined here, we need to determine the absorption at long wavelength where $\alpha \leq 10^{-3}$ cm^{-1}. Since the measured absorption spectra (Fig. 2) are often noisy at $\alpha \sim 10^{-3}$ cm^{-1},

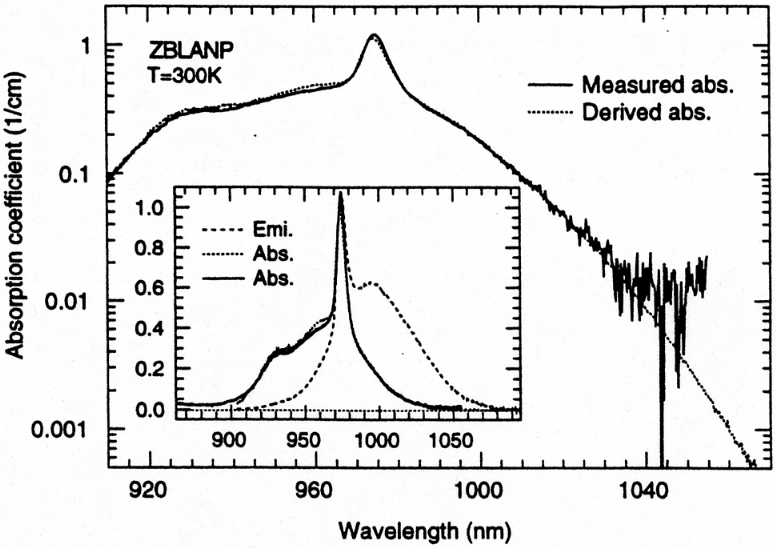

Fig. 7. A comparison of measured and reciprocity-derived absorption spectra of ZBLANP. The inset are linear plots of the measured (solid line) and derived (doted line) absorption spectra and the measured emission (dashed line) inthe cross section (10^{-20} cm^2).

we used the absorption derived from the emission spectrum by reciprocity method, developed by McCumber [22], [23] and generalized by Band [24] and Payne [25]. In terms of the absorption coefficient and the emissivity per unit wavelength, the reciprocity relation can be written as

$$\alpha(\lambda) \propto \lambda^5 \varepsilon(\lambda) \exp[hc/\lambda kT] \tag{2}$$

which is valid as long as the ground and excited states are each in thermal equilibrium with the host. Fig. 7 shows a plot of the room-temperature absorption coefficient α as measured and as derived by the reciprocity relation from the emission spectra of ZBLANP; the two spectra normalized at 1010 nm are in good agreement. The reciprocity relation allows us to determine the absorption coefficient in the long-wavelength cooling tail with higher precision.

C. Radiative Life-Times

Following [26], we derived the radiative lifetime τ_{rad} from the measured absorption coefficient, using

$$\tau_{\text{rad}} = \frac{3N\overline{\lambda}_{\text{abs}}^{-4}}{32\pi cn \int \alpha(\lambda)\, d\lambda} \tag{3}$$

where $\overline{\lambda}_{\text{abs}}$ is the mean absorption wavelength, N is the Yb^{3+} site density, and n is the refractive index. The radiative lifetimes obtained by (3) are 1.6 ms for BIGaZYT, 1.7 ms for ZBLANP and 2.0 ms for QX/Yb phosphate glass. A shorter radiative lifetime would benefit the cooling power. Table II summarizes spectroscopic properties of Yb^{3+}-doped ZBLANP, BIGaZYT, and QX/Yb phosphate glasses at room temperature.

D. Cooling Efficiency

Each fluorescent photon carries off, on average, thermal energy equal to the difference between the pump-photon and the mean fluorescent-photon energies. When nonradiative relaxation from the excited to the ground state is negligible, the cooling power P_{cool} is proportional to the absorbed pump power P_{abs} and to the average difference in the photon energies of the pump and fluorescence radiation. The cooling efficiency is thus given by

$$\eta(T) = P_{\text{cool}}/P_{\text{abs}} = [\lambda_p - \overline{\lambda}(T)]/\overline{\lambda}(T) \tag{4}$$

where λ_p is the pumping wavelength. In the design of practical optical refrigerators, the cooling material is pumped in the cooling tail where $\alpha \sim 10^{-5}$–10^{-3} cm^{-1}. To compare the potential cooling of the three glasses being examined here, we find the pumping wavelength λ_p corresponding to $\alpha \sim 10^{-3}$ cm^{-1} and compute the cooling efficiency for all temperatures. Since the measured absorption spectra (Fig. 3) are often noisy at $\alpha \sim 10^{-3}$ cm^{-1}, we used the absorption derived by reciprocity, as described above. The cooling efficiency as a function of temperature estimated by (4) is shown in Fig. 8. The lines are the least-square fits to data with an exponential function $a(T - T_{\min}) \exp(-bT)$, which is empirically found to provide a good characterization of the temperature-dependent behavior of the cooling efficiency. This figure suggests that Yb:BIGaZYT has the potential to

TABLE II
SPECTROSCOPIC PROPERTIES OF YB^{3+} IN ZBLANP, BIGAZYT, AND QX PHOSPHATE GLASSES

Properties	units	ZBLANP	BIGaZYT	QX/Yb
Mean fluorescence	$\overline{\lambda}(nm)$	995	1005	1004
Mean absorption	$\overline{\lambda}_{abs}(nm)$	963	961	959
Radiative life-time	$\tau_{rad}(ms)$	1.7	1.6	2.0
Abs. cross-section	$\int \alpha(\lambda)d\lambda(10^{-3})$	3.8	71	6.2
Phonon energy	$\hbar\omega_{max}(cm^{-1})$	580	430	1100

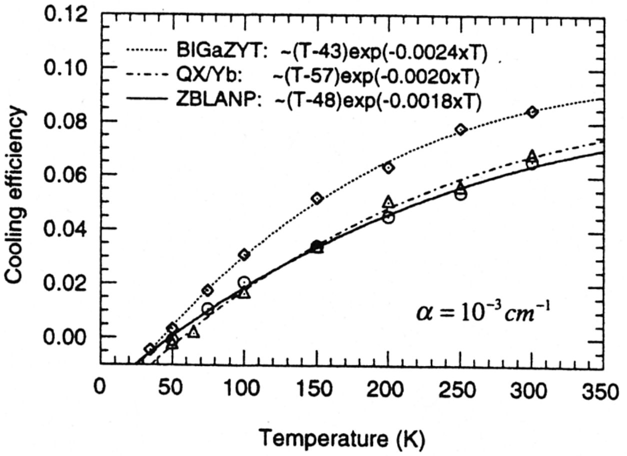

Fig. 8. The cooling efficiency as a function of temperature of the ZBLANP, BIGaZYT, and QX/Yb phosphate glasses.

be significantly better than Yb:ZBLANP, which is currently the only glass with reported cooling [18]. This is largely determined by the Yb^{3+} site densities in the hosts, which were ~1.1, 2.2, and 20 ($\times 10^{20}$ cm^3) for ZBLANP, QX/Yb, and BIGaZYT, respectively (see Table I). In particular, the figure indicates BIGaZYT glass could have a cooling efficiency more than twice that of ZBLANP at temperatures below 80 K and may attain a minimum temperature (at zero load) of 45 K compared to 55 K expected for the ZBLANP.

IV. Conclusions

The spectroscopic properties of Yb^{3+}-doped ZBLANP, BIGaZYT, and QX phosphate glasses have been evaluated to assess their value as optical refrigerants. The assessment is based on the shape of the cooling tail of the absorption spectra and the mean fluorescence wavelength as functions of temperature. BIGaZYT glass has the possibility of cooling more than twice as efficiently as ZBLANP at temperatures below 80 K, largely due to its high tolerance of Yb^{3+} dopant concentration. QX/Yb phosphate glass possesses a high thermal conductivity which would be an asset in its integration into a practical cooler.

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